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The aims of this program were to synthesize increasingly high-quality thin film and single crystals of the high Tc and other novel superconductors. We also proposed to study their physical properties, understand the nature of their superconductivity, and search for new superconducting devices and device concepts, with a primary objective of developing a better understanding of the limits of the occurrence of superconductors with high transition temperatures.

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Final Technical Report
due 31 October, 1994

Principal Investigators
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February 28, 1994

Final Technical Report

AFOSR Contract F49620-92-C-0004

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I. INTRODUCTION

The goals of the Stanford program in this project were to synthesize increasingly high-quality thin films and single crystals of the high-T_c superconductors and other novel superconductors, to study their physical properties, understand the nature of their superconductivity and search for new superconducting devices and device concepts.

A primary objective of the program was to develop a better understanding of the limits of the occurrence of superconductors with high transition temperatures (above liquid nitrogen temperature). Thus, in this work we include parts that reflect our search for new materials and new ways to synthesize them, as well as measurements that provide us with critical tests of theories and potential applications. This last possibility also led us to device model systems that include the main physical ingredients of high Tc materials to study some specific problems. An example is the employment of artificially layered superconductor/insulator systems of MoGe/Ge which have in-plane short coherence length and large penetration depth, and out of plane controlled coupling that allow us to produce similar anisotropies to existing high-Tc materials.

II. ACCOMPLISHMENTS

Following the above introduction, we divide our accomplishments into three categories. The first is Materials. Here we discuss new approaches and success in growing new materials, as well as progress in the development of new tools to synthesize materials. The second category is the study of fundamental properties that relate to potential use of high-Tc materials. The last category is the direct study of high-Tc devices or device concepts.

Materials

1. The "Infinite Layer" Material

Using our MBS system we have deposited thin films of the so-called infinite layer material $SrCuO_2$ and the normally conducting perovskite $SrRuO_3$ that has shown potential as a normal barrier material in high-Tc SNS Josephson devices. Preliminary doping studies of the infinite layer material have been undertaken. *In situ* UPS studies of the ruthenate show a very large density of states at the Fermi level, raising interesting questions about the origin of the high resistivity of this material.

2. Sr Doped Fullerene Sr_xC_{60}

Under the AASERT supported part of our program, we have explored the materials science and electronic structure of the Sr doped fullerene Sr_xC_{60}, with x = 0.2 to 6. Unlike previous workers, we are doping the material using codeposition, as opposed to post-deposition diffusion of the dopant from the surface. In principle, our approach should lead to more uniform doping. The fullerene source is a temperature stabilized Knudsen cell, and the Sr dopant was deposited using an electron beam heated source and atomic absorption rate control. Preliminary evidence shows differences in the valance band photoemission spectra from our codeposited films and those reported in the literature doped via diffusion. These possible differences are under continuing investigation.

3. The surface of $YBa_2Cu_3O_7$

We have demonstrated that the surface of YBCO thin films can be cleaned by means of a thermal anneal in the presence of an atomic oxygen beam. The cleaned surfaces yield well-developed LEED patterns and a Fermi edge in photoemission spectroscopy at room temperature. Previously, photoemission studies were only possible on surfaces cleaved at cryogenic temperatures. Studies of SNS Josephson junctions on these surfaces are underway.

4. Development of Atomic Absorption Rate Control for Advanced Vapor Phase Synthesis of Films

Working with Intelligent Sensor Technology Inc, a small instrumentation company in Silicon Valley, we have developed a new approach to atomic absorption rate control that defines the state of the art. We have achieved 1% noise at a deposition rate of 0.3 Å/sec with a control bandwidth suitable for atomic layer-by-layer growth using electron beam heated sources. This has been achieved in the high background pressures of activated oxygen needed for

growth of the cuprate superconductors. The approach appears to have potential utility for sputtering processes used in semiconductor manufacturing.

In addition, we also have submitted an STTR proposal with Intelligent Sensor Technology, Inc., to continue developing hollow cathode lamp atomic absorption systems. Under the auspices of the Center for Materials Research here at Stanford, we have also combined forces with the nonlinear optics group at Stanford and New Focus, Inc., in an ARPA proposal to develop tunable diode laser atomic absorption systems. This proposal led to a recently awarded grant.

Fundamental Properties

1. Structural and Transport Properties of Grain Boundaries

We have investigated the structure and properties of well-defined model grain boundaries, being motivated by the fact that grain boundaries are important in Josephson junctions and in high-field high-current conductors.

The transport properties of three types of 90-degree grain boundaries were compared using (103) oriented $YBa_2Cu_3O_7$ (YBCO) thin films grown epitaxially on (101) $SrTiO_3$ and (101) $LaAlO_3$ substrates. The films were grown using the *in situ* 90-degree off-axis sputtering approach developed earlier by our group. The in-plane crystallographic film orientation is given by the YBCO <301> parallel to the substrate [101]. A domain structure exists with the CuO_2 planes oriented at $\pm$ 45 degrees to the substrate surface (i.e., parallel to the substrate [010] direction). Specific sets of 90 degree grain boundaries are observed in both principal in-plane directions. The normal-state conductivity and the critical current density of these films along the YBCO [010] direction are as high as the best quality c-axis films, which have no high-angle grain boundaries. This demonstrates that twist boundaries (Type A in Fig. 1) have no discernible weak link nature. The normal-state conductivity and critical current density along the <301> direction are much lower than in the [010] direction. The normalized magnetic-field dependence of J_c for both those directions is similar and shows no evidence of weak link behavior. The anisotropic transport behavior in the normal and superconducting state can be explained by the microstructure and a simple transport model.

2. Transport Properties of $PrBa_2Cu_3O_7$

A-axis-oriented $YBa_2Cu_3O_7/PrBa_2Cu_3O_7$ superlattices and their transport properties have been grown with smooth surface and high crystalline quality. The materials can be visualized as a parallel array of narrow 1-D-like channels of the copper oxide planes of YBCO separated by the PBCO layers. The magnetic field dependence of the resistive transitions of superlattices containing individual $YBa_2Cu_3O_7$ layers 24 or 48 Å thick, separated by $PrBa_2Cu_3O_7$ layers

24 or 48 Å thick, shows an apparent dimensional cross-over at a temperature T* that depends on the $PrBa_2Cu_3O_7$ thickness. Above T* the transition is insensitive to magnetic fields (up to 8 T); below T* some broadening occurs. These results indicate an abrupt disappearance of the coupling between the $YBa_2Cu_3O_7$ channels in the structure above T*.

3. Properties of High-Tc and Artificially Layered Materials in Magnetic Field

A universal feature of the high-Tc cuprate superconductors is their strong anisotropy. This anisotropy fundamentally affects the properties of these superconductors in the vortex state. We are studying the effects of such anisotropy both by studying the cuprates directly (such as in the a-axis YBCO/PbCO multilayers described above) and by using a novel model system based on the artificially-structured MoGe/Ge superconductor/insulator multilayer system with anisotropies that can be adjusted to match those of high-Tc superconductors. We are also studying for comparison the behavior of individual layers, an approach that is not yet possible in the high-Tc materials.

3.1 Irradiation Effects on the Irreversibility Line of $Bi_2Sr_2CaCu_2O_8$

More specifically, we have studied the defect dependence of the irreversibility line in $Bi_2Sr_2CaCu_2O_8$ for magnetic fields along the c-axis. We find that the irreversibility line of pristine single crystals exhibit three regimes. For fields less than 0.1 T, it obeys a power law, $H_{irr} = H_0(1-T_{irr}/T_c)^{\mu}$, where μ and H_0 are functions of T_c. For fields greater than 2 T, the irreversibility line becomes roughly linear ($\mu = 1$) with a slope of 0.7 T/K. For intermediate fields, there is a crossover region, which corresponds to the onset of collective vortex behavior. Defects produced by proton irradiation shift the irreversibility line in all three regimes. The high-field regime moves to higher temperatures, the low-field regime moves to lower temperatures, and the crossover to collective behavior becomes obscured. A maximal increase in the irreversibility temperature in the high-field regime is found to occur at a defect density of nearly one defect per vortex disk. These results demonstrate that the high-field current carrying capacity of 2212 BSCCO, which is limited by the position of the irreversibility line, can be increased by the introduction of appropriate (columnar) defects.

3.2 MoGe as a Model System

To better understand the relation between transport and thermodynamic transitions in high-Tc superconductors we compared the resistivity, magnetization and specific heat of a superconducting amorphous $Mo_{77}Ge_{23}$ layer separated by insulating amorphous Ge. The fluctuation magnetization and specific heat are in excellent agreement with the 2D lowest Landau level scaling

approximation. The broadening of the resistive transition is similar to that observed in the specific heat, but it is not in quantitative agreement with the proposed scaling relations. The resistivity becomes activated immediately below the thermodynamic transition, indicating little or no region that is describable by conventional flux flow.

Using our thin Mo-Ge films we have also studied the resistive transition of two-dimensional superconductors (i.e., single layers) in a magnetic field. Activation energies for vortex motion as a function of magnetic field have been deduced from the data. Study of the systematics of these data convincingly shows that at low fields an edge barrier to vortex entry governs the resistance. The relevance of this effect for superconducting flux flow transistors will be interesting to examine. At higher fields and for the thinnest films, we find that the activation energy decreases logarithmically with increasing magnetic field. This same behavior has been reported in c-axis YBCO/PBCO multilayers. Thus we believe we are examining the same vortex dynamics as arise in the high-Tc materials. The data are consistent with recent theories which predict that at high J_c the activation energy for flux motion is governed by motion of dislocations in the vortex lattice. For thicker films, which have lower critical current densities, a non-monotonic field dependence is seen that empirically correlates with the theoretical field dependence expected for vortex motion in the form of correlated "flux bundles" with a diameter given by the Larkin-Ochinokov correlation length. Thus, both of these behaviors arise for film thicknesses less than those for which the vortex melting was observed. The precise connection between these two regimes needs to be elucidated but clearly reflects the lower critical current densities of the thicker films.

We have also used measurements of the ac penetration depth of amorphous MoGe multilayers and single films in the presence of a perpendicular magnetic field to study the nature of the correlations of the vortex lattice of highly anisotropic and two-dimensional superconductors. The results reveal an anomaly in the ac response of the vortex lattice at a characteristic temperature below the $H_{c2}(T)$ line. We have found that the field and frequency dependence of this anomaly is consistent with a Kosterlitz-Thouless type melting of the two-dimensional vortex lattice on short length scales. However, we observe a crossover in the frequency dependence which suggests that even below the melting temperature the vortex lattice remains disordered on long length scales. This is in fact the first direct and unambiguous observation of a Kosterlitz-Thouless-type melting of the vortex lattice in any kind of superconductor. Moreover, it confirms the suspicion that many previous claimed observations (in particular those for high-Tc superconductors) were of electromagnetic origin (i.e., skin-depth effects) and not actually indicative of melting.

4. Perpendicular Transport in BSCCO Crystals in the Presence of a Magnetic Field

We studied the c-axis transport in single crystals of BSCCO. The data suggest that decoupling of the superconducting CuO_2 layers occurs via a continuous crossover in this material. Both zero bias resistance and I-V measurements were taken along the transition. The resistance along the c-direction continues its normal state trend, although the ab plane has already started its superconducting transition. At lower temperatures the resistance along that direction peaks and then starts to decrease rapidly. This decrease then slows down to a usual exponential tail. We find experimental evidence that the initial interlayer couplings appear in the field-dependent temperature regions of the maxima in the c-axis resistivity. Most of the c-axis resistance drops in a temperature range between the peak and the foot, while the rest of the resistance vanishes in a much slower fashion, presumably all the way to the irreversibility line. We also performed Monte Carlo simulations of decoupling which properly include in-plane as well as interlayer interactions between pancake vortices, and predicts that interlayer phase coherence will be established across all of the layers of a crystal at field-dependent temperatures which are in excellent agreement with the positions of the feet of our c-axis resistive transitions.

5. Magnetic Field Dependence of the Specific Heat of High-Tc Superconductors—Evidence for Lines of Nodes

The question observing low lying excitations due to nodes in the gap of non- simple s-wave superconductors has been a controversial one. The main reason was that any additional contribution observed at low temperatures could always be attributed to dirt effects since specific heat cannot distinguish the type of excitations. In particular, the ability to observe the effect of possible nodes due to a $d_{x^2-y^2}$ symmetry in high-Tc materials was a great challenge. Recently, we employed a new technique that overcomes these difficulties. Since a magnetic field affects only the charged degrees of freedom, it is possible to distinguish the density of states of the charged excitations if we measure their magnetic field dependence, and in particular along different orientations. Such a technique was used by us to determine the electronic density of states at the Fermi level, $N(E_F,H)$ for $YBa_2Cu_3O_{6.95}$ single crystals made at the University of British Columbia. We have found that the total specific heat is best described by including two predictions for the electronic specific heat of d-wave superconductivity: a T^2 term in zero field and an increased linear term in a magnetic field applied perpendicular to the CuO_2 planes. The additional linear term, which implies a finite $N(E_F,H)$ µ $(H/H_{c2})^{1/2}$ was also predicted by G. Volovik for superconductivity with lines of nodes in the gap. Following our experiment, and using similar analysis, Ramirez *et al.* showed that the specific heat of heavy electron superconductors behaves in a similar way.

6. Magnetic Properties of $SrRuO_3$

$SrRuO_3$ is a potential material for the "normal" part in SNS junctions involving high-Tc superconductors. Such junctions were already fabricated at Conductus Inc. with very promising characteristics. In order to further study the properties of this system which is inherently ferromagnetic below 160 K, epitaxial thin films of $SrRuO_3$ were fabricated by the group of Kookrin Char at Conductus Inc. Films were measured with a bulk magnetometer and with a local magneto-optic Sagnac interferometer in transmission and in reflection. We found a magnetic easy axis perpendicular to the film, and for saturated magnetization along this direction the Faraday rotation and the Kerr rotation at λ=840 nm are $0.75\text{x}10^5$ deg/cm and 0.85 deg, respectively. The temperature dependence of the magnetization in the low temperature limit is dominated by spin-wave excitations, yielding notable decrease with a $T^{3/2}$. Using Sagnac-Kerr scanning and TEM imaging we were able to correlate the coercivity of these films with the grain size.

Device Concepts

1. Feasibility of the Flux Flow Transistor

I-V characteristics of low-T_c, amorphous Mo-Ge alloy flux flow transistors have been carried out collaboratively with Jon Martens of Conductus. The high sheet resistance, low critical-current densities, and well-known materials parameters of the Mo-Ge system make it ideal for testing the device potential of low-T_c flux flow transistors. No flux flow transistor action was obtained. The I-V curves were dominated by self-heating effects, except very near to T_c. Even near T_c, the I-V curve of the device could only be modulated (due to thermal effects) when the control line was driven into the normal state. An analysis of these devices using well-known theories of heating in superconducting thin-film strips confirmed the importance of heating.

2. Thermal Boundary Resistance as a Limiting Factor for Bolometers

Under the AASERT-supported part of our program, we studied the difference in thermal boundary resistance between c-axis and a-axis films. In general, one would like to minimize this resistance to obtain better thermalization between the film that acts as a sensor and the substrate. The inability to thermalize them results in a longer response time of the device. Our experiments showed that the thermal boundary resistance between $YBa_2Cu_3O_7$ and $LaAlO_3$ substrates are 6 times lower for a-axis films than for c-axis films. This result strongly suggests the use of a-axis films in bolometer applications. In addition, we found that the thermal boundary resistance of a-axis films drops a small factor right at Tc.

3. Possible Josephson Junctions with $Bi_2Sr_2CaCu_2O_8$

Josephson junctions using Pb counter electrodes have been made in both the c-axis and a-axis directions of single crystal BSCCO. Josephson coupling was observed only in the a-axis direction. The results are relevant in the debate on the symmetry of the pair wave function in the cuprate superconductors. Taken at face value, they are consistent with (but do not prove) d-wave pairing. In addition, previous anomalies in the magnetic diffraction patterns of the a-axis junctions have now been tentatively explained as being due to vortex penetration into the lower BSCCO electrode of the junction. Vortex penetration into the BSCCO is a reflection of the very low H_{c1}'s of the cuprate superconductors for fields along the ab plane. Vortex penetration into the electrodes of high-T_c Josephson junctions will require modification of the usual device models of these devices.

III. VISITORS TO OUR PROGRAM

An important ingredient of our program is to interact with other groups working on similar subjects as well as with industry. In particular we are interested in transferring our knowledge and experience to industrial partners. This goal is primarily achieved by attracting visitors to Stanford. The list below reflects this interaction.

Short Term Visitors

Dr. K. Nakamura	National Inst. of Metals	Nov. 11–24, 1991
M. Kupriyanov	Moscow State Univ.	Jan. 2-5, 1992
Dr. Imafuku	Nippon Steel Corp.	April 16, 1992
Dr. Ray Ashoori	AT&T	April 22, 1992
Dr. Hans Mooij	Delft	April 26–27, 1992
Dr. David Nelson	Harvard University	April 28, 1992
Dr. Gun Yong Song	ETRI	May 4, 1992
Ken Daley	TRW	Sept. 17, 1992
Alfred Lee	TRW	Sept. 17, 1992
Claire Pittiet-Hall	TRW	Sept. 17, 1992
Marc Gabay	Orsay Univ. , Paris, France	July Aug., 1992
Melissa Charalambous	IBM, Yorktown	Nov. 1994
Marc Gabay	Orsay Univ. , Paris, France	July Aug., 1993
Marc Gabay	Orsay Univ. , Paris, France	July Aug., 1994
Matthew Fisher	U.C. Santa Barbara	May, 1994

Long Term Visitors

Toshimasa Umezawa	Yokogawa Electric Co.	June 1991–Dec. 1992
Alberto Pomar	Universidad de Santiago	Jan. 1992–Feb. 1992
Dr. Guntherodt	Univ. Basel, Switzerland	July 1–Aug. 16, 1992
John Clem	Iowa State University	Sept. 1992–July 1993
Roland Busch	Siemens AG	Oct.–Dec. 1992
Zafer Durusoy	Hacettepe University	Oct. 92–May 1993
Phillipe Fluckiger	Univ. Neuchatel, Switzerland	Dec. 1993 - Nov. 1994
Patrick Fournier	Univ. Scherbrook, Canada	Jan. 1994 - present
Lior Klein	Bar-Ilan Univ. , Israel	Aug. 1993 - present

IV. GRADUATE STUDENTS RECEIVING THEIR PhD UNDER THIS PROGRAM

Steven Spielman	Lawrence Berkeley Labs Berkeley, CA	1992
Debra Jean Lew	Princeton University Princeton, NJ	1994
Jeffrey Stuart Urbach	University of Texas Austin, TX	1993
Louis Lombardo	Adelphi Technology Palo Alto, CA	1994
Whitney Ryan White	AT&T Bell Labs Holmdel, NJ	1994
Seungoh Ryu	Ohio State University Columbus, OH	1994
Ali Yazdani	IBM Almaden Research Center San Jose, CA	1994

V. OTHER OUTREACH PROGRAMS

Our group attracts not only graduate students and visitors but also tries to help with other educational programs within and outside Stanford. At Stanford we attract a few interested undergraduate students who either come to work in our group as a summer job, or, stay to do an honors thesis. In the past three years we had about 10 undergraduate students contributing at different levels to our program.

In addition, we hosted Professor Khilstrom from Westmont College, an undergraduate school at Santa Barbara. Professor Khilstrom used his time here to prepare samples and devices to be used in his research and teaching at his institution.

VI. PUBLICATIONS UNDER THIS PROGRAM

1. Debra Lew, Yuri Suzuki, C. B. Eom, M. Lee, J.-M. Triscone, T. H. Geballe, and M. R. Beasley, "Vertical Transport Properties of a-Axis Oriented YBa_2Cu_3O/ $PrBa_2Cu_3O_7$ $YBa_2Cu_3O_7$ Sandwich Junctions," *Physica C* **185-189** (1991) pp. 2553-2554 (North Holland.)

2. M.R. Beasley, "Tunneling and Proximity Effect Studies of the High Temperature Superconductors," *Physica C* **185-189** (1991) pp. 227-233 (North Holland).

3. M.R. Beasley, W.R. White, M. Hahn, and A. Kapitulnik, "Vortices in Artificially-Structured Quasi Two-Dimensional Superconductors," *Physica Scripta*, **T42**, 25-28, 1992. (Invited paper, presented at Jubilee Nobel Symposium, December 4-7, 1991, Goteborg, Sweden.)

4. Debra Lew, Yuri Suzuki, C. B. Eom, M. Lee, J.-M. Triscone, T. H. Geballe, and M. R. Beasley, "Vertical Transport Properties of a-Axis Oriented $YBa_2Cu_3O_7$/ $PrBa_2Cu_3O_7$/$YBa_2Cu_3O_7$ Sandwich Junctions," *Physica C*, **185-189** (1991)pp. 2553-2554 (North Holland).

5. M. R. Beasley, "Tunneling and Proximity Effect Studies of the High Temperature Superconductors," *Physica C* **185-189** (1991) pp. 227-233 (North Holland).

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7. L.W. Lombardo, D.B. Mitzi, A. Kapitulnik, and A. Leone, "Defect dependence of the irreversibility line in $Bi_2Sr_2CaCu_2O_8$ single crystals," *Phys. Rev. B* **46**, 5615 (1992).

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